Lightweight High Temperature Alloys based on the AlFeSi System

PI: Dr. Michele Manuel, Co-PI: Dr. Richard Hennig

University of Florida

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Project ID: mat069

Overview

Timeline

- Start Data: October 01, 2016
- End Data: December 31, 2019
- Percent Complete: 70%

Budget

- Total Project Cost: $1,102,082
  - DOE share: $991,873
  - Non-DOE share: $110,209
- Funding received in FY17: $342,611
- Funding received in FY18: $320,194
- Funding for FY19: $329,068

Project partners

GM | Oak Ridge National Laboratory | University of Nebraska Lincoln | Illinois, University of Illinois at Urbana-Champaign

Barriers

- **Weight:** Light-weight materials are needed for vehicle weight reduction in order to meet the future more stringent fuel economy standards
- **Cost:** Low-cost aluminum alloys with high-temperature properties are currently not available
- **Fabrication:** The τ_{10}-$Al_4Fe_{1.7}Si$ phase has an extremely small composition range and a small fluctuation in composition will change the solidification path creating an unwanted microstructure

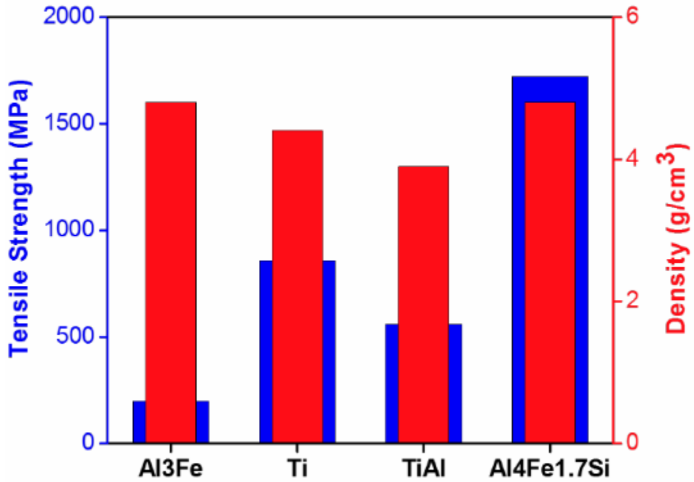
Objectives & Relevance

Project Objective:

- To develop low-cost lightweight τ_{10}-$Al_4Fe_{1.7}Si$ based alloys that meet or exceed the high-temperature performance of more expensive titanium aluminides and Ni-based superalloys by combining alloy chemistry strategies and 3D additive manufacturing technologies

Relevance to the Barriers:

- The high strength-to-weight ratio of τ_{10} makes it ideal for high temperature, lightweight applications
- All the three base constituents are available abundant in nature and low cost
- Additive manufacturing enable high cooling rate and can produce components to exact shape with no additional machining



General Motors (GM):
US Patent 2017/0211168 A1

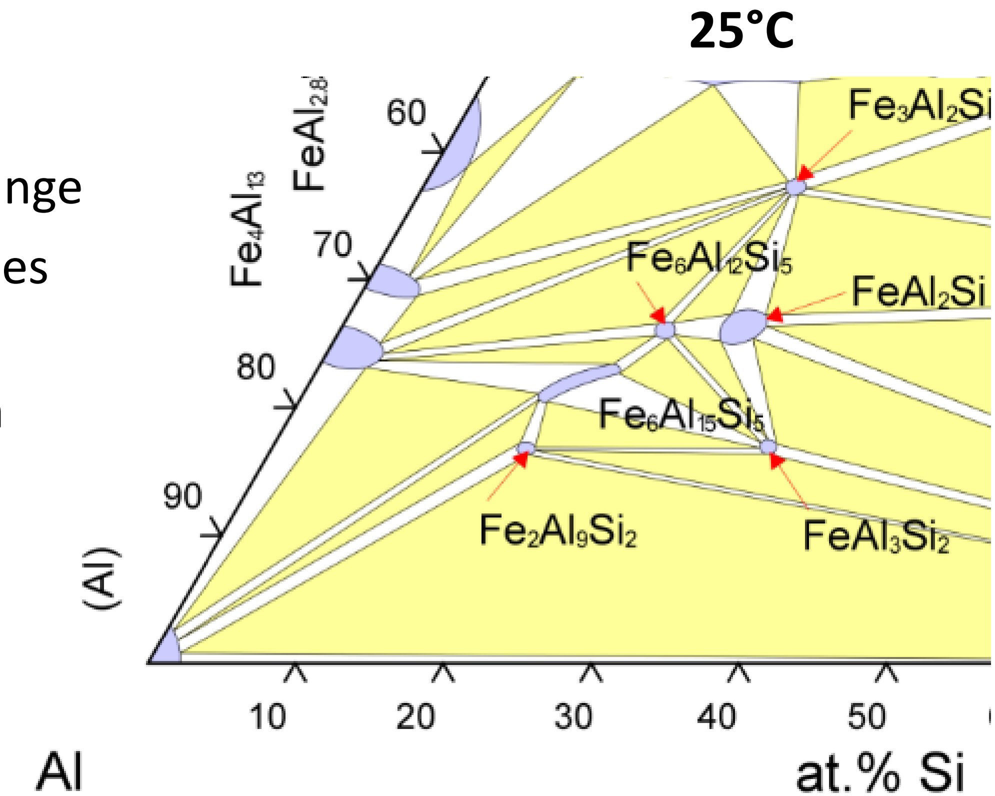
Relevance

Difficulties:

- ❑ Extremely limited composition range
- ❑ Large number of competing phases at high temperature
- ❑ No longer a stable phase at room temperature (25 °C)

Needs either:

- ❑ Phase stabilization
- ❑ Non-equilibrium solidification



ASM Phase Diagram Center

Technical strategies:

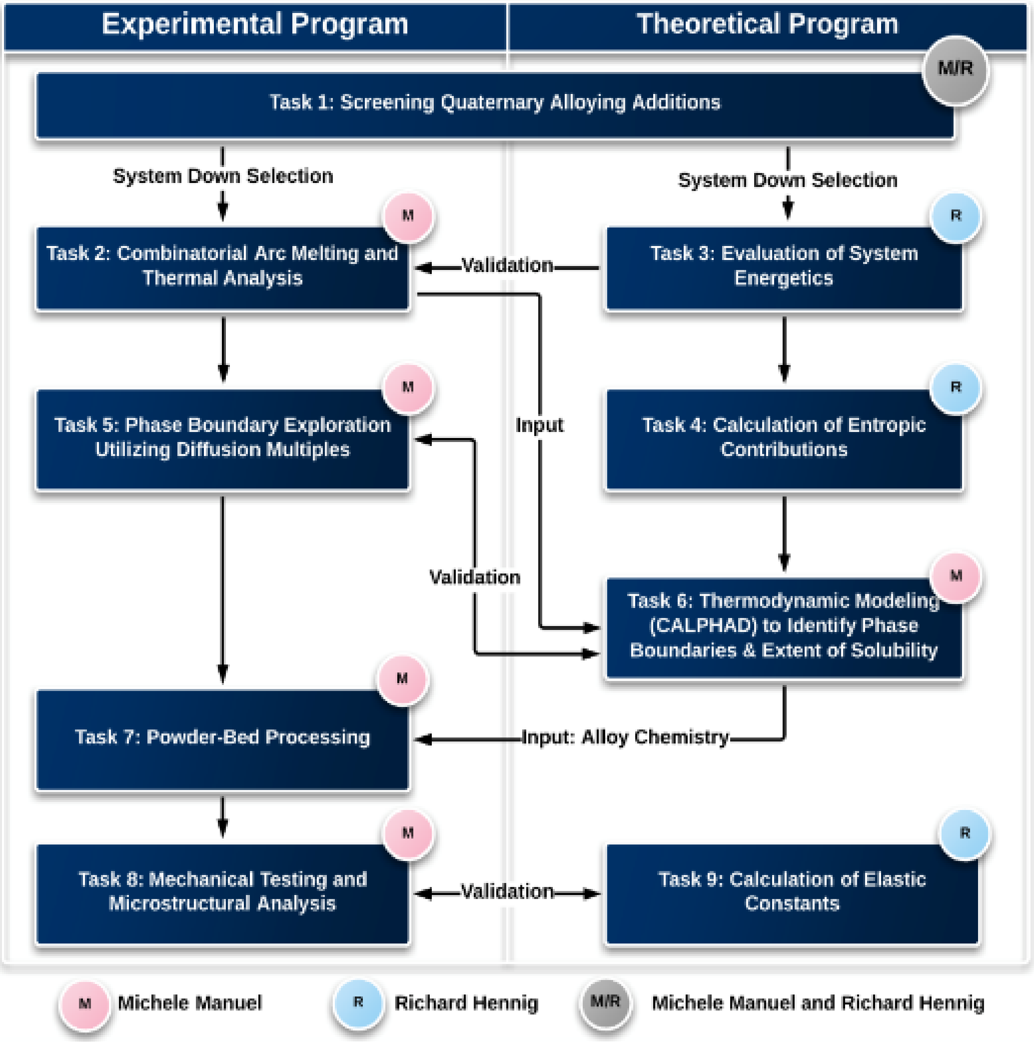
- ❑ Expansion of the composition range by addition of minor alloying elements
- ❑ High cooling rate enabled by 3D laser printing

Milestones

Budget	Milestone	Planned Completion	Actual Completion	Percent Completion
Period 1 FY17	**Quaternary systems down-selected (Technical):** Identify potential quaternary solute elements that increase the composition range	Oct-17	Oct-17	100%
	Quaternary systems fabrication complete (Technical): Creation of arc melted buttons that have the potential to display a stable τ_{10}-Al-Fe-Si-X phase	Oct-17	Dec-17	100%
	Initial alloys delivered (Go/No Go): Deliver alloys that have the potential to display a stable τ_{10}-Al-Fe-Si-X phase	Oct-17	Dec-17	100%
Period 2 FY18	**Database of compound thermodynamics developed (Technical):** Calculation and evaluation of solute components that stabilize the phase of interest while destabilizing competing phases	Oct-18	Dec-18	100%
	Solution modeling of phase stability ranges at high and low temperatures (Technical): Experimentally-validated thermodynamic models will be created that explain each the high temperature solubility and low temperature metastability in quaternary systems of interest	Oct-18	Dec-18	100%
	Chemistry list drafted (Go/No Go): Creation of a list of chemistries with good high-temperature phase stability and suitable for powder processing via 3D printing	Oct-18	Dec-18	100%
Period 3 FY19	**Production of powders (Technical):** Production of powders in the quaternary systems of interest	Dec-19		30%
	Production of additively manufactured components (Technical): Production of mechanical test samples using additive manufacturing	Dec-19		
	Calculation of elastic constants (Technical): Database of elastic constants for systems of interest completed	Dec-19		
	Ability to additively manufacture parts (Go/No Go): Additively printed parts must be mechanically stable enough to test for tensile, compression and fatigue properties	Dec-19		

Approach/Strategy

- The overall objective of this project is to combine alloy chemistry and 3D laser printing to create phases with unusual properties
- Scope includes both computational and experimental work, culminating in producing 3D laser sintered components for mechanical testing

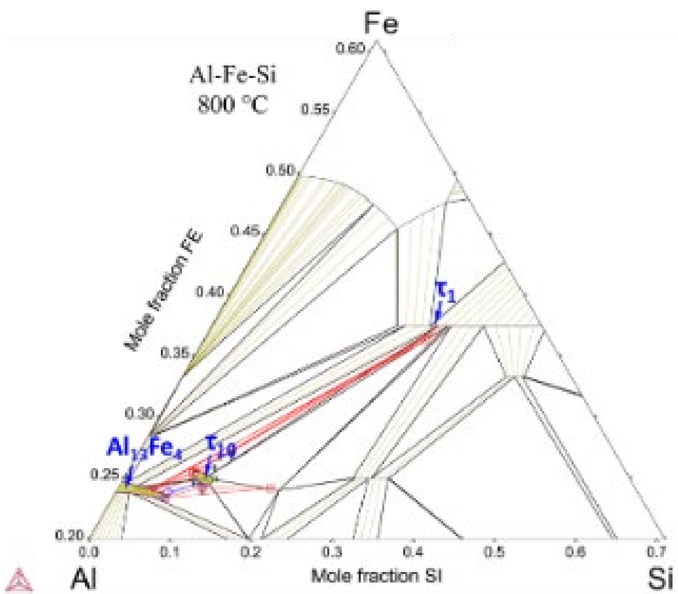


Technical Accomplishments

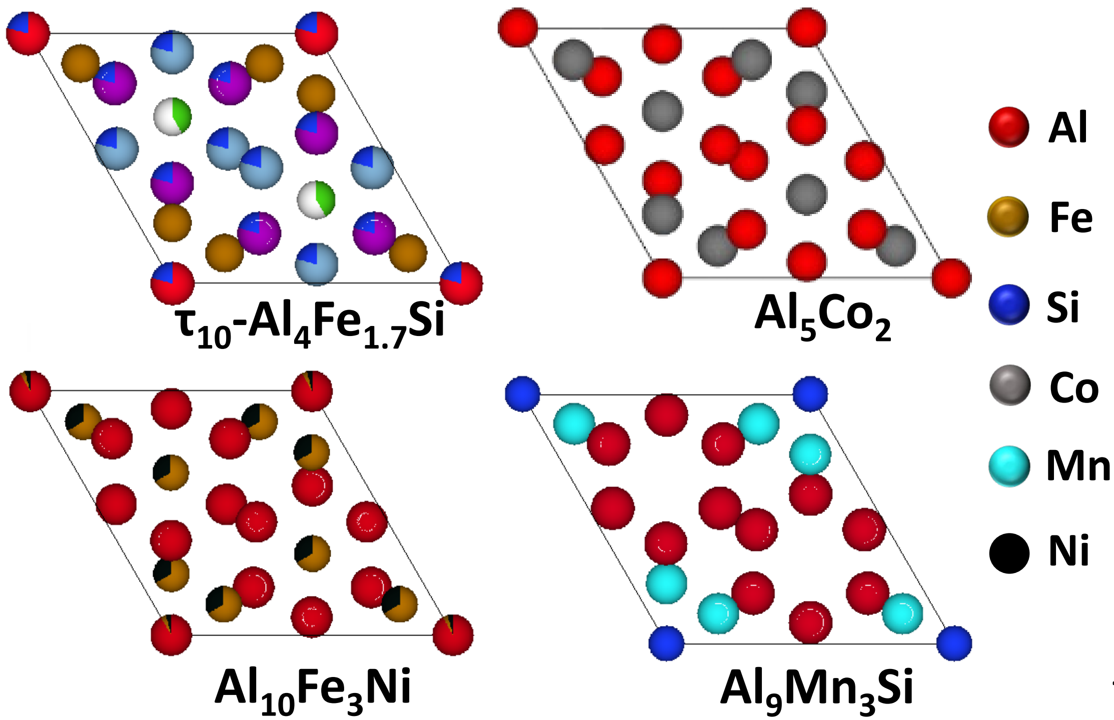
A brief review of the Tasks 1.0 & 2.0 in FY17

A sorted list of elements that enhances the stability of the τ_{10} phase

- The stable compositional range of τ_{10}-$Al_4Fe_{1.7}Si$ at 800 & 950 °C were measured, which are wider than the thermodynamic calculations
- Co, Ni, Mn and Zn were selected as promising quaternary additions by data-mining of crystal structure databases



Al-(24.3-25.5)Fe-(8.2-10.8)Si



Technical Accomplishments

Task 3.0 – Evaluation of system energetics

Calculations for chemical substitutions of alloying elements in the τ_{10} phase to determine candidates for addition

- Ni, Ti, and Co are below the hull; expected to be soluble in the τ_{10} phase
- Cu and Zn are close and could be stabilized by entropy of mixing
- Ni, Co, Cu, and Zn could substitute on Si site, and Ti could substitute on Al site
- **Ni, Ti, Co, Cu and Zn are predicted to be promising candidates for alloying in the τ_{10} phase**

Quaternary Element	Crystal Structure Site	Hull Distance (eV)	Quaternary Element	Crystal Structure Site	Hull Distance (eV)
Nb	Al	5.53874	Ti	Al	-0.11475
	Fe	3.51976		Fe	-0.06046
	Si	0.03564		Si	0.00475
Cu	Al	4.33047	Mo	Al	5.70977
	Fe	3.04199		Fe	3.56938
	Si	0.01018		Si	0.03439
Zn	Al	5.34495	Co	Al	4.91206
	Fe	2.86860		Fe	3.21411
	Si	0.01619		Si	-0.00840
Ni	Al	4.64192	Sn	Al	4.38951
	Fe	3.12342		Fe	3.11125
	Si	-0.01437		Si	0.05859

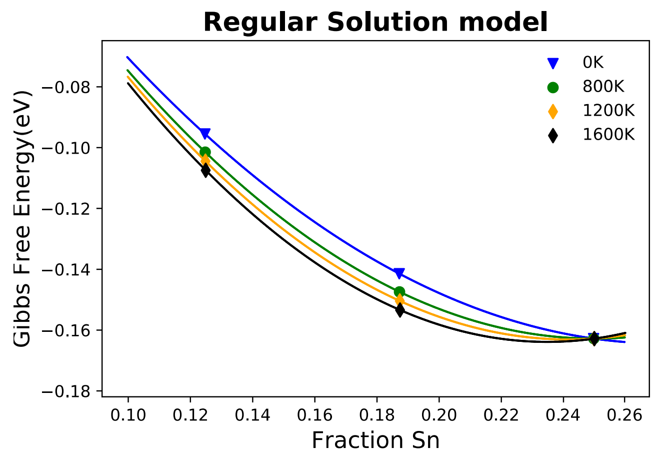
The hull distance of quaternary compound (AlFeSi-X). The configurations were formed replacing either Al, Fe or Si in the τ_{10} phase with the quaternary element in order to check the solubility of the element

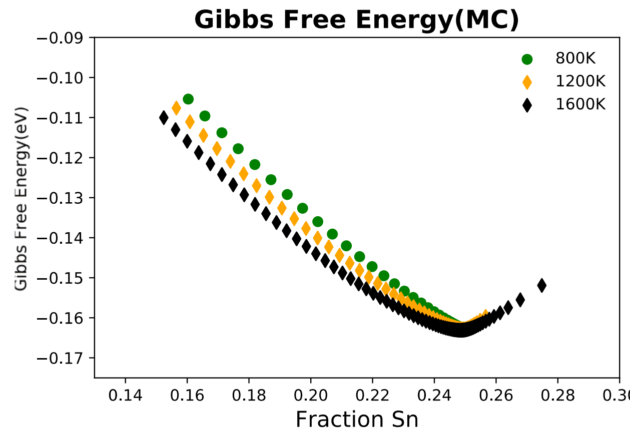
Technical Accomplishments

Task 4.0 – Calculation of entropic contributions

Develop methodology to estimate entropic stabilization of solubility

- In order to develop methodology, as a proof of concept to calculate the entropic contribution to energy, we chose Nb-Sn binary system
- Two approaches to calculate entropy were chosen:
  1) Regular solution model
  2) Cluster expansion of DFT energies and Monte Carlo integration
- Found that regular solution model provides reasonable estimates and is computationally efficient for multi-component systems
- Next: Apply the cluster expansion and Monte Carlo to τ_{10} and competing phases for entropic contribution



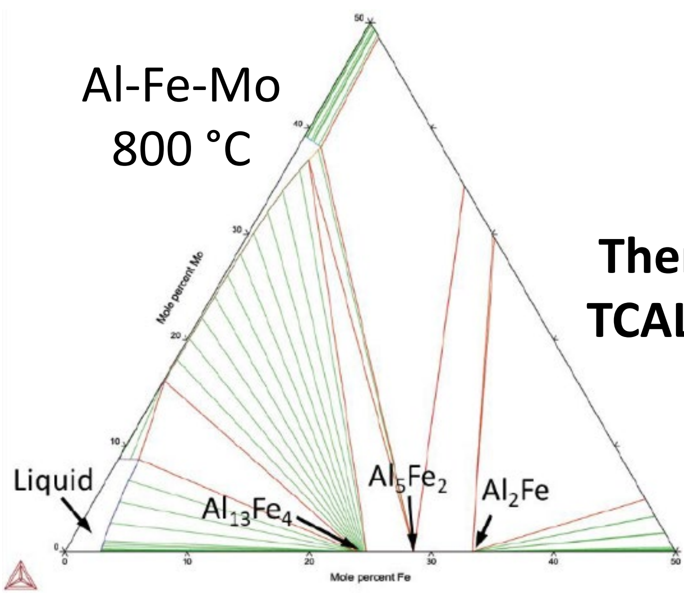


Technical Accomplishments

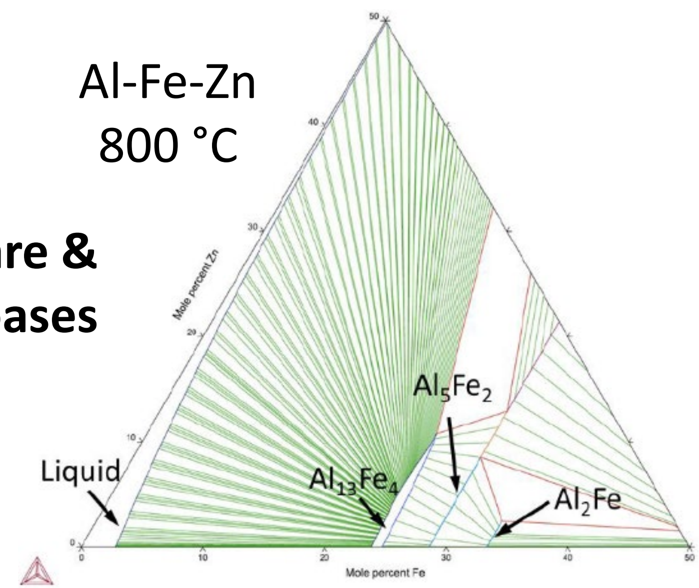
Task 6.0 – Thermodynamic modeling

Calculated phase diagram to predict the solubility of quaternary alloying additions in the competing $Al_{13}Fe_4$ phase

- $Al_{13}Fe_4$ is the main competing phase in almost all τ_{10} alloys
- Hypothesis: element with a low solubility in the competing $Al_{13}Fe_4$ phase has the potential to destabilize it
- The calculations guided the diffusion couple experiments in Task 5.0



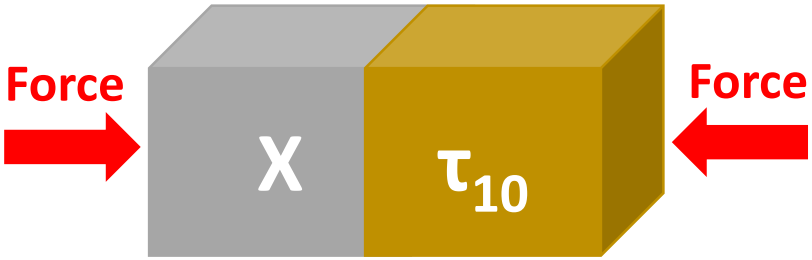
Thermo-Calc software & TCAL5 / TCFE9 databases



Technical Accomplishments

Task 5.0 – Phase boundary exploration: diffusion couple

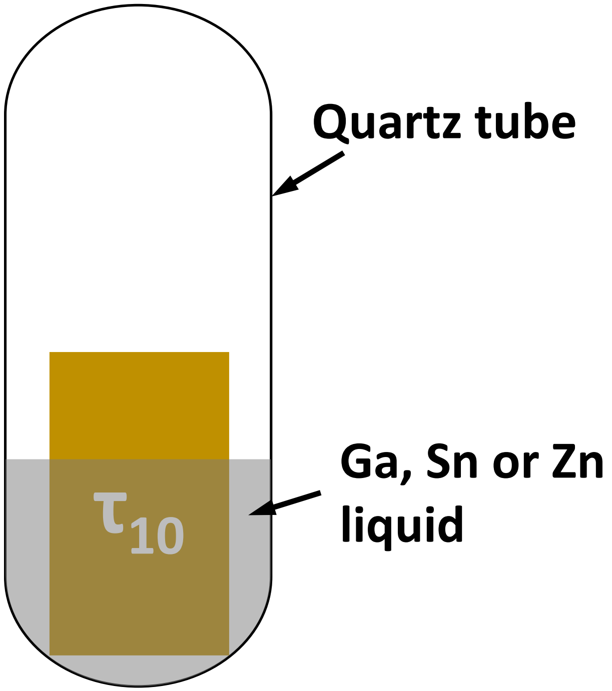
Exact solubility limit of the quaternary solutes in the τ_{10} phase



Solid-solid diffusion couple (SSDC)

τ_{10}/X (X= Co, Cu, Mn, Mo, Nb, Ni, Ti)

Diffusion couple jig (Kovar alloy)



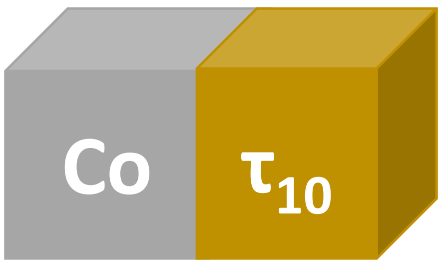
Liquid-solid diffusion couple (LSDC)

τ_{10}/X (X = Ga, Sn, Zn)

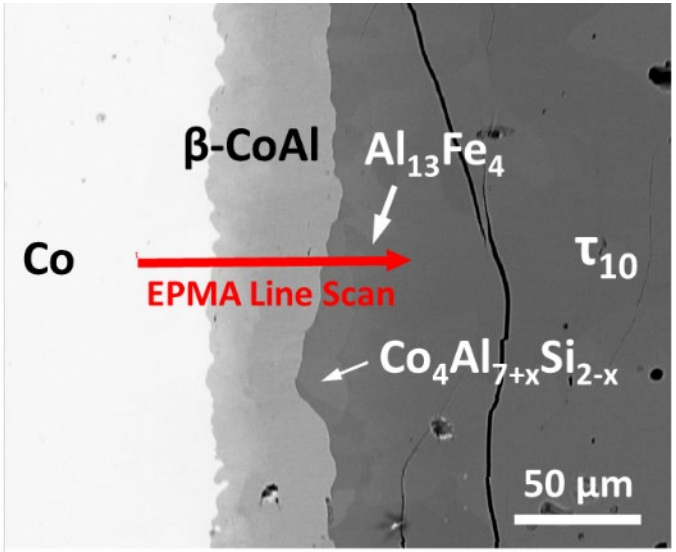
Technical Accomplishments

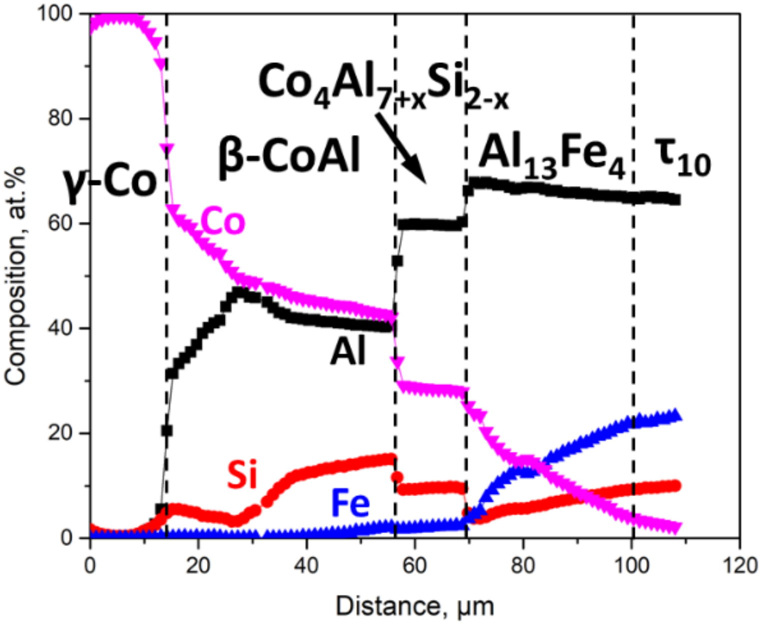
Task 5.0 – Phase boundary exploration: τ_{10}/Co SSDC

An example of SSDC for measuring the solubility of Co in τ_{10}



τ_{10}/Co SSDC
800 °C, 336 h



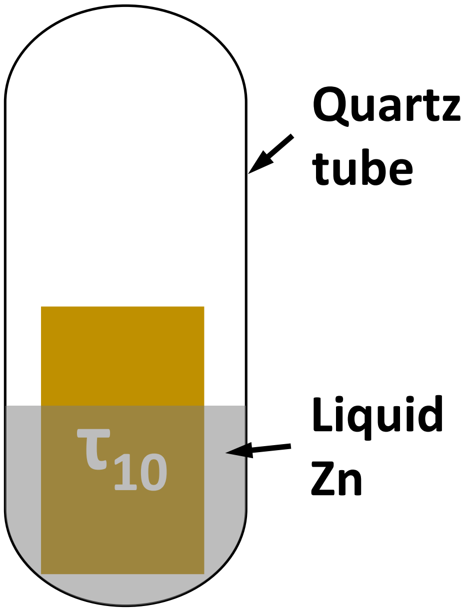


Solubility of Co in τ_{10} was determined to be 3.5 at.% at 800 °C

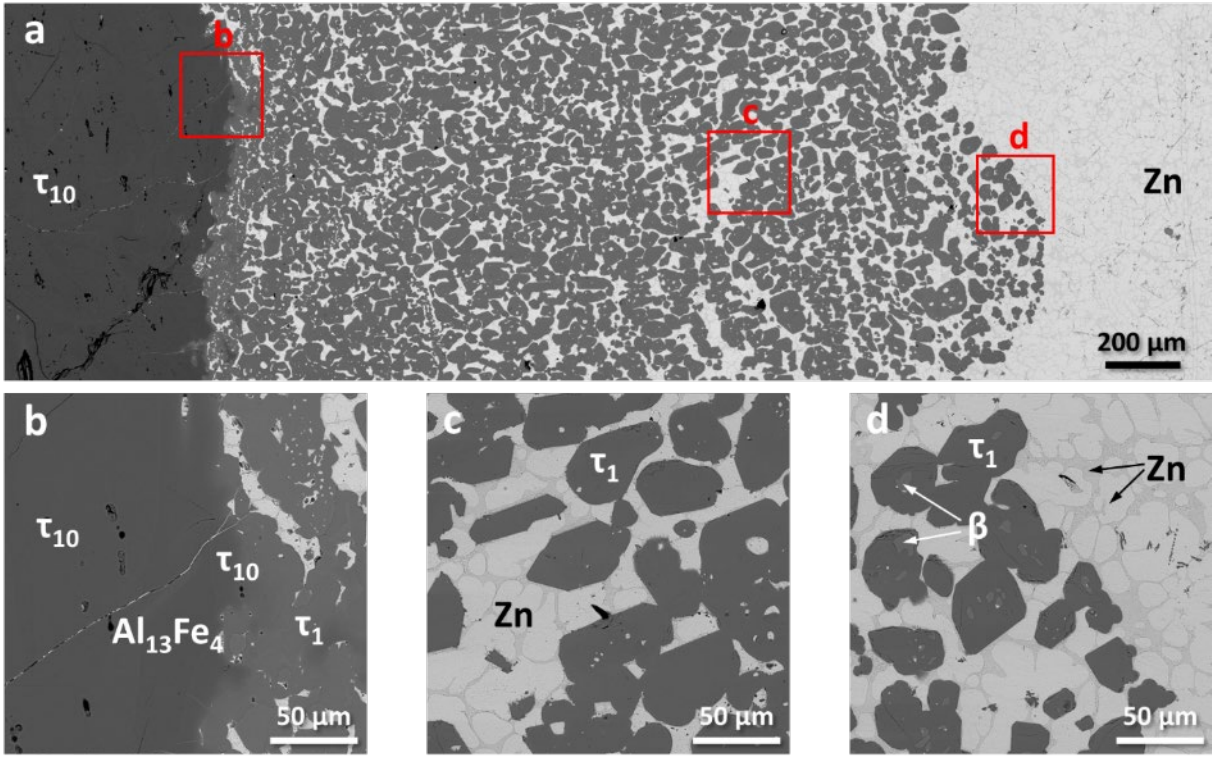
Technical Accomplishments

Task 5.0 – Phase boundary exploration: τ_{10}/Zn LSDC

An example of LSDC for measuring the solubility of Zn in τ_{10}



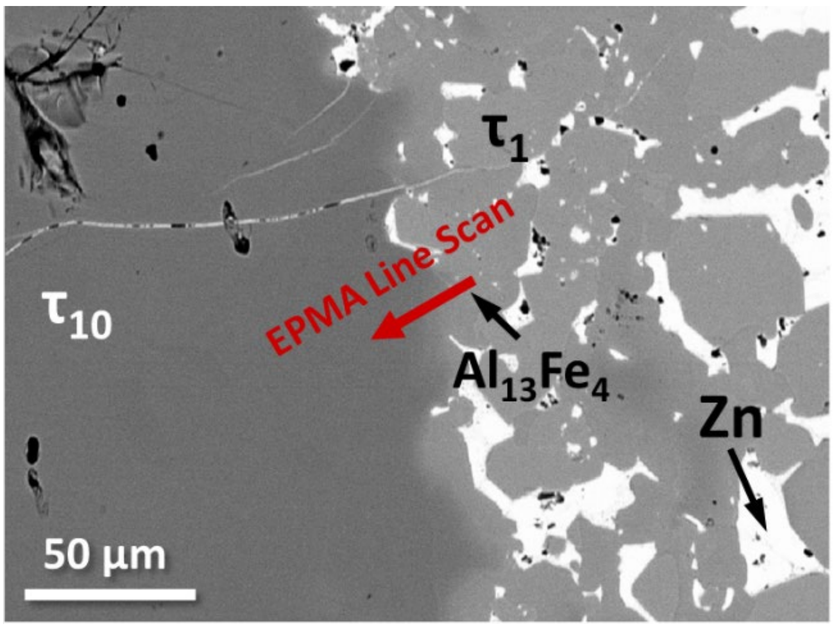
τ_{10}/Zn LSDC
800 °C, 10 h

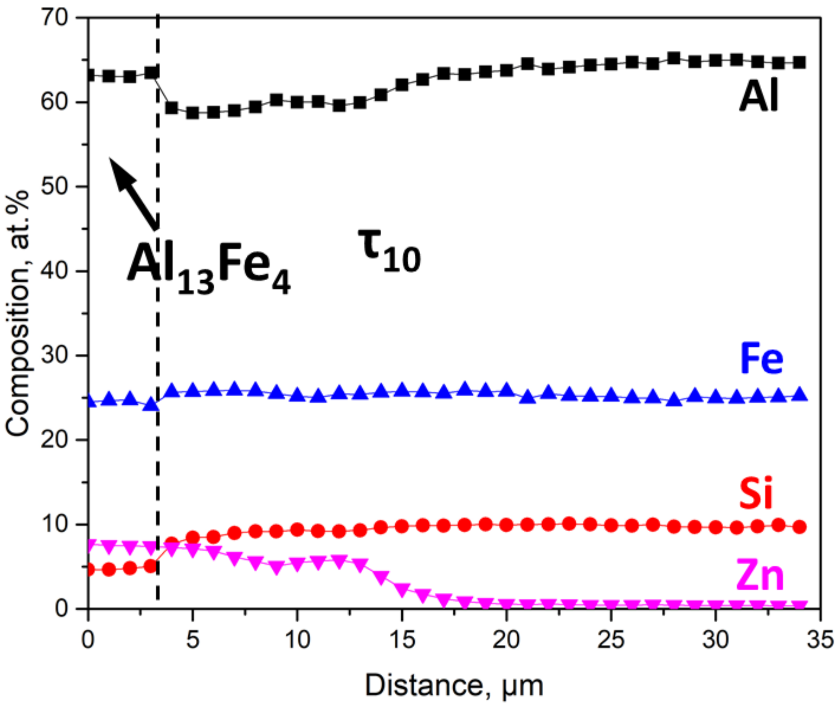


Technical Accomplishments

Task 5.0 – Phase boundary exploration: τ_{10}/Zn LSDC

An example of measuring the solubility of Zn in τ_{10}

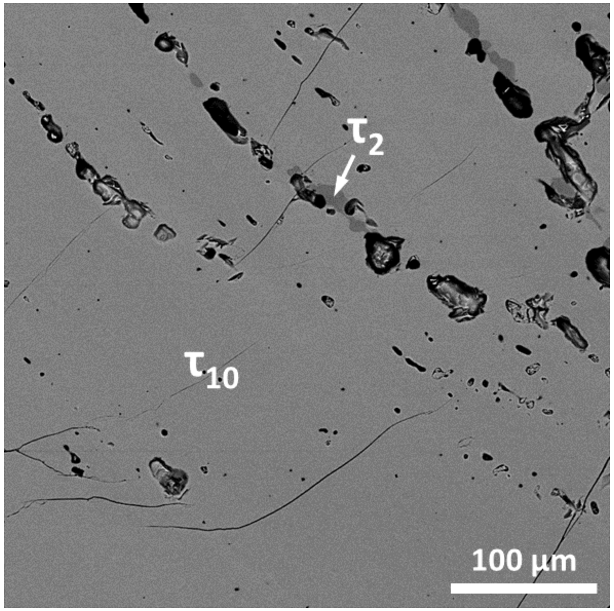




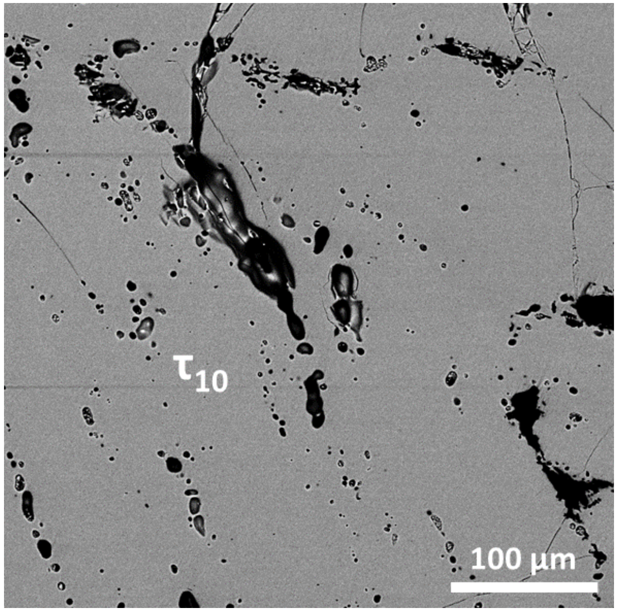
- **Solubility of Zn in τ_{10} is relatively high, up to 7.2 at.% at 800 °C**
- **Zn primarily replaces Al position in τ_{10}**

Technical Accomplishments

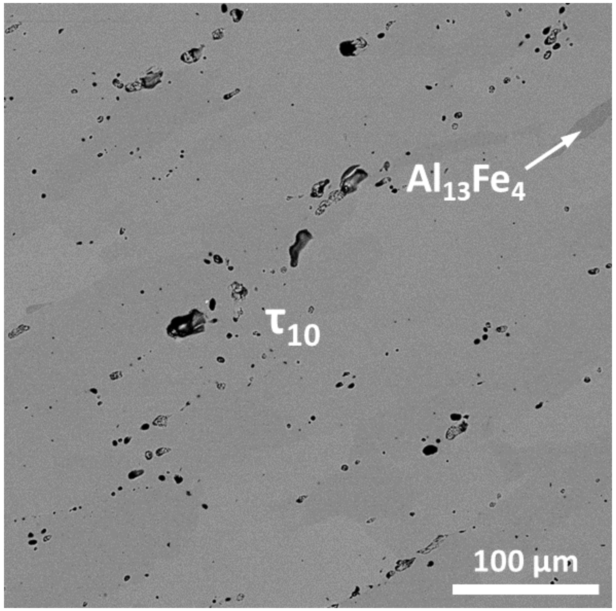
Task 5.0 – Phase boundary exploration: alloy verification



Al-23.0Fe-8.0Si-2.3Cu
800 °C, 500 h



Al-20.0Fe-11.0Si-4.5Mn
800 °C, 400 h



Al-24.2Fe-8.0Si-2.0Ni
800 °C, 500 h

- **Three quaternary alloys were confirmed to have good stability of the τ_{10} phase**
- **Powders are being created for additive manufacturing process**

Responses to Previous Year Comments

Comment	Response
The weakness lies in the generality of some of the areas being suggested such as creating a list of chemistries as a go/no-go point. One could create a list of chemistries while sitting at a desk that could be argued meets the objective of being producible or printable; it is the specific claims behind this list that would be compelling.	The initial chemistry list was done based on the results from diffusion couples and casted alloys. This is just a preliminary list that is being used to fabricate the first batch of powders for 3D printing. The list will be adjusted depending on the results obtained from the powder production and printed parts. And in addition, AlFeSiX alloys are still being studied.
Definition of more specific materials properties targets, including targeted operating temperatures and strengths at those temperatures.	Base on input from the industry we are targeting tensile strengths greater than 450MPa and phase stability at temperatures up to 800 °C with a strength of 400MPa.
The elastic constants predicted in task 9 will be single crystal constants, while the measurements performed in task 8 will likely be on highly oriented polycrystal samples. The reviewer asked what will be a successful validation in this case.	The calculated elastic constants will guide the experimental efforts to identify optimal solute concentrations for experiments in Task 8. The results from experimental testing will in turn guide the calculations. It will act as a feed back loop between computation and experiments rather than validating each others.

Partnerships and Collaborations

GM

Provided induction melted ingots and has participated in monthly and annual onsite meetings. The continually offer advice and recommendations on solute selection and alloy design

Oak Ridge National Laboratory

Worked with beamline scientist to study site-lattice occupancy of elements within the τ_{10}-$Al_4Fe_{1.7}Si$ phase. This information will be used to validate computational models

University of Nebraska Lincoln

Will start additive manufacturing process to fabricate AlFeSiX components for mechanical testing

Illinois – University of Illinois at Urbana-Champaign

Provided feedback on the study of site-lattice occupancy of elements within the τ_{10}-$Al_4Fe_{1.7}Si$ phase

Remaining Challenges and Barriers

- High thermodynamic stability of competing phase, such as $Al_{13}Fe_4$, add an extra layer of complexity as additional solute elements should stabilize the τ_{10}-$Al_4Fe_{1.7}Si$ phase while de-stabilizing detrimental competing phases
- The τ_{10}-$Al_4Fe_{1.7}Si$ phase is a high temperature phase which is only stable between 727 and 997 °C. The high cooling rate (~10^7 K/s - ~10^{13} K/s) enabled by 3D laser printing to selectively promote the solidification of τ_{10} single phase. Therefore, the proper processing conditions for 3D laser printing need to be explored to control the solidification path without creating an unwanted microstructure
- Considering the brittleness and processing difficulties of the τ_{10}, the additive manufacturing will be used to fabricate the net shape components for mechanical testing. So, to effectively understand and mitigate residual stress effects on the additive manufactured parts will remain a challenge

Proposed future work

- Continue to expand the phase boundaries of the τ_{10}-$Al_4Fe_{1.7}Si$ intermetallic phase with the addition of minor quaternary alloying elements
- Obtain more AlFeSiX chemistries that have good high-temperature phase stability and are suitable for powder processing via 3D printing
- Produce AlFeSiX powders and fabricate components by 3D printing for microstructural analysis and mechanical testing
- Calculate elastic constants for quaternary τ_{10} phases

Ongoing	FY19	**Q4 Milestone (Task 7) — Production of powders:** Production of powders in the quaternary systems of interest
		Q4 Milestone (Task 8) — Production of additively manufactured components: Production of mechanical test samples using additive manufacturing
		Q4 Milestone (Task 9) — Calculation of elastic constants: Database of elastic constants for systems of interest completed
		Q4 Go/No Go Decision Point — Ability to additively manufacture parts: Additively printed parts must be mechanically stable enough to test for tensile, compression and fatigue properties

Any proposed future work is subject to change based on funding levels.

Summary

Project Objective:

- To develop low-cost lightweight τ_{10}-$Al_4Fe_{1.7}Si$ based alloys that meet or exceed the high-temperature performance of more expensive Ni-based superalloys and titanium aluminides by combining alloy chemistry strategies and 3D additive manufacturing technologies

Results:

Budget Period 1

- The stable compositional range of the τ_{10}-$Al_4Fe_{1.7}Si$ at 800 and 950 °C
- Co, Cu, Mn, Ni and Zn were selected as promising quaternary additions to expand the composition range of τ_{10}-$Al_4Fe_{1.7}Si$
- Two Al-Fe-Si-Ni alloys were obtained with the single τ_{10} phase

Budget Period 2

- Identified ten potential quaternary alloying additions and determined their solubility in τ_{10}
- Validated method for estimating entropy contribution of alloying elements
- Fabricated AlFeSi-Co, AlFeSi-Cu, AlFeSi-Mn, and AlFeSi-Ni quaternary alloys with high phase volume of the τ_{10} stable phase
- Obtained two alloy chemistries for powder fabrication and then 3D printing
- Experimental data were collected in order to improve the current Al thermodynamic databases for designing AlFeSi-based alloys

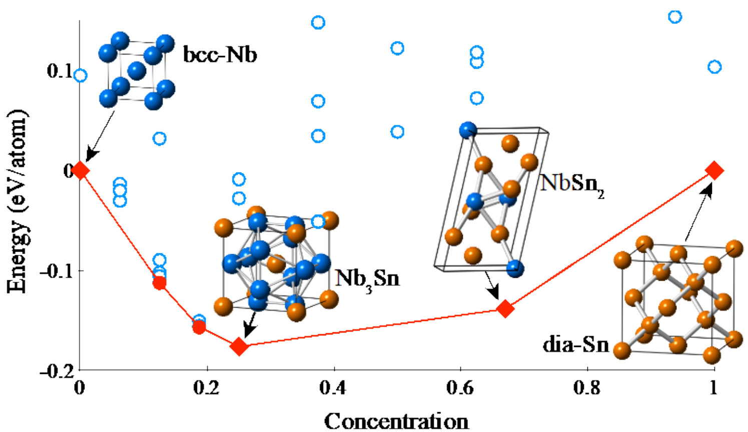
Technical Back-up Slides

Technical Accomplishments

Task 4.0 – Calculation of entropic contributions

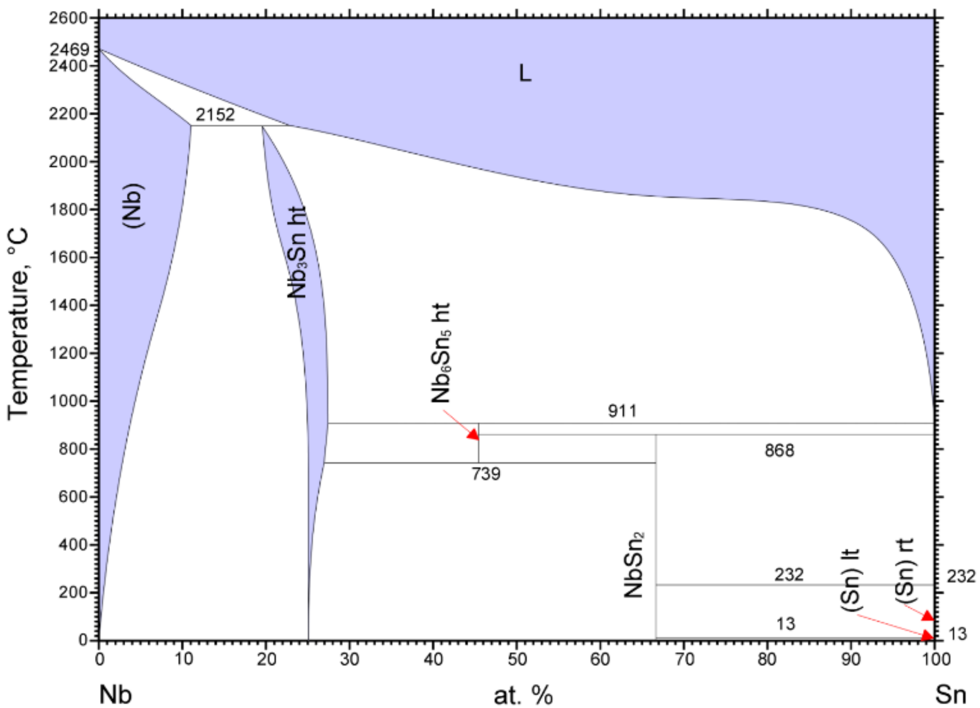
Energetics of the binary Nb-Sn system

- Intermetallic A15 Nb_3Sn phase shows asymmetric solubility
- Structure search identifies low-energy defects that control the solubility



However, our results show that solubility of Nb in Nb_3Sn phase at lower temperature

ASM Phase Diagram Center



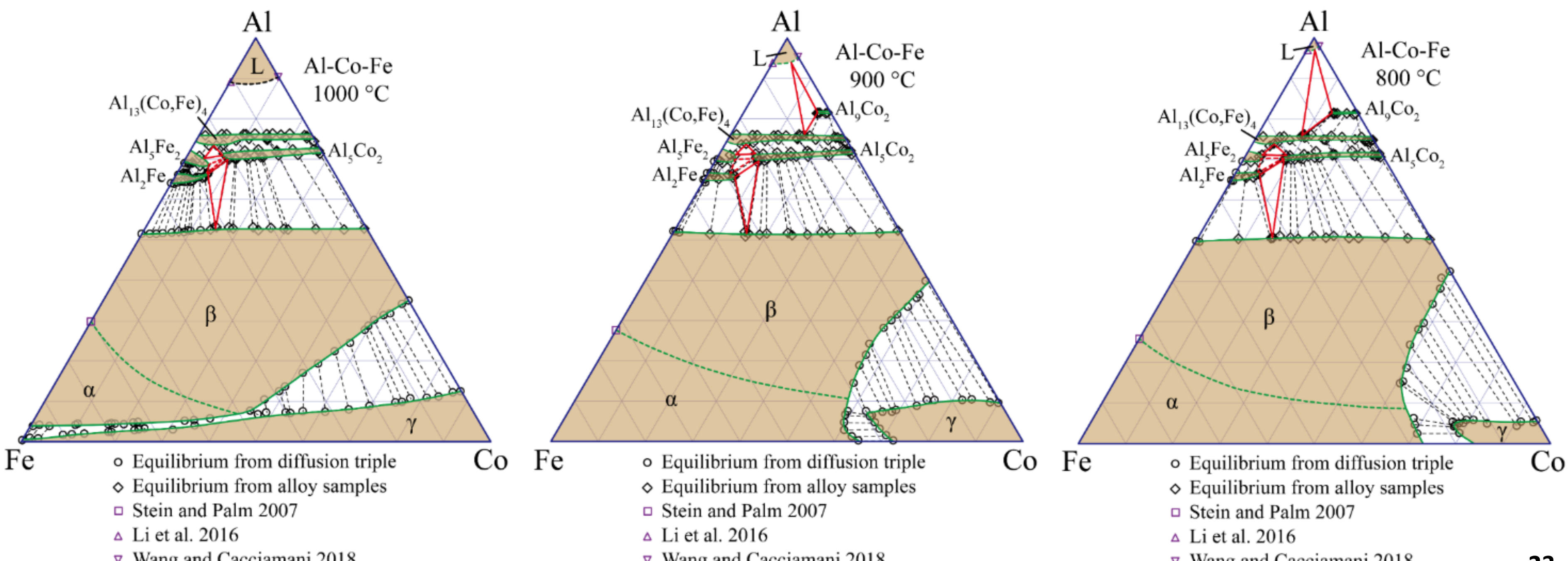
ASM phase diagram showing Nb_3Sn as a line compound at lower temperatures

Task 6.0 – Thermodynamic Modeling

Collect more experimental data for future thermodynamic assessments to improve the current Al thermodynamic databases for designing AlFeSi-based alloys

- Determine the Al-Fe-Co and Al-Fe-Cu phase diagrams over the whole composition range
- Investigate phase equilibria and diffusion in the Fe-Zn system

Al-Fe-Co phase diagram over the whole composition range



Task 6.0 – Thermodynamic Modeling

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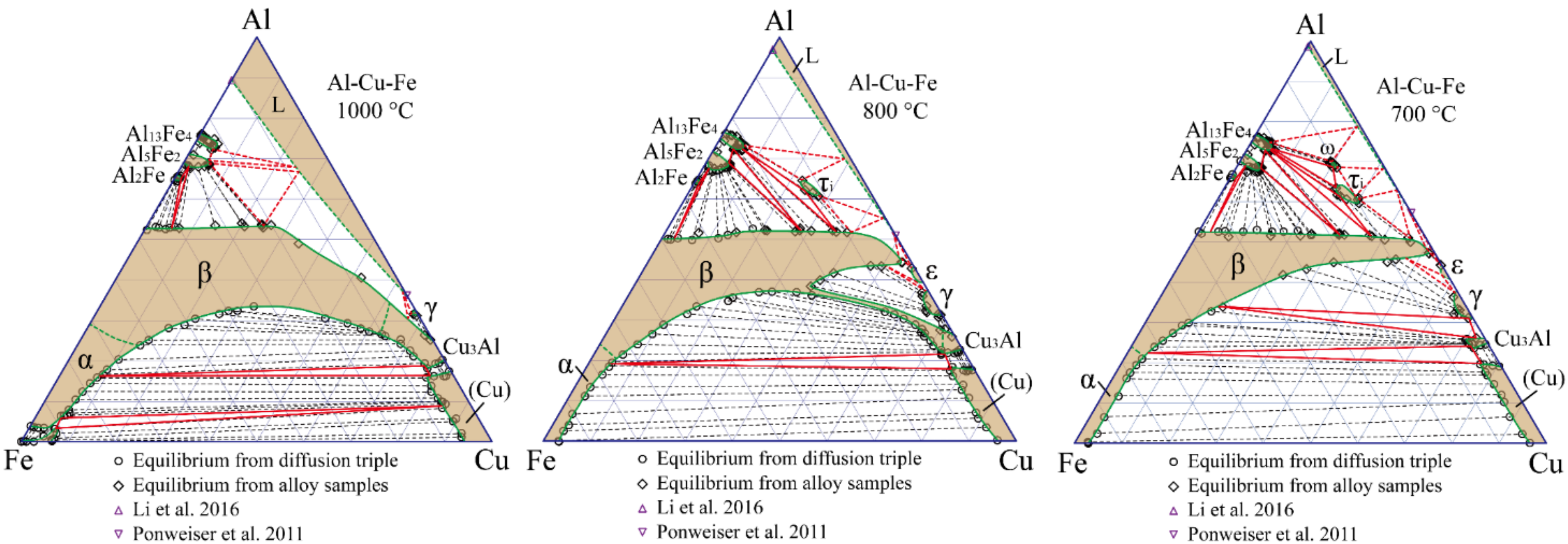
Al-Fe-Cu phase diagram over the whole composition range

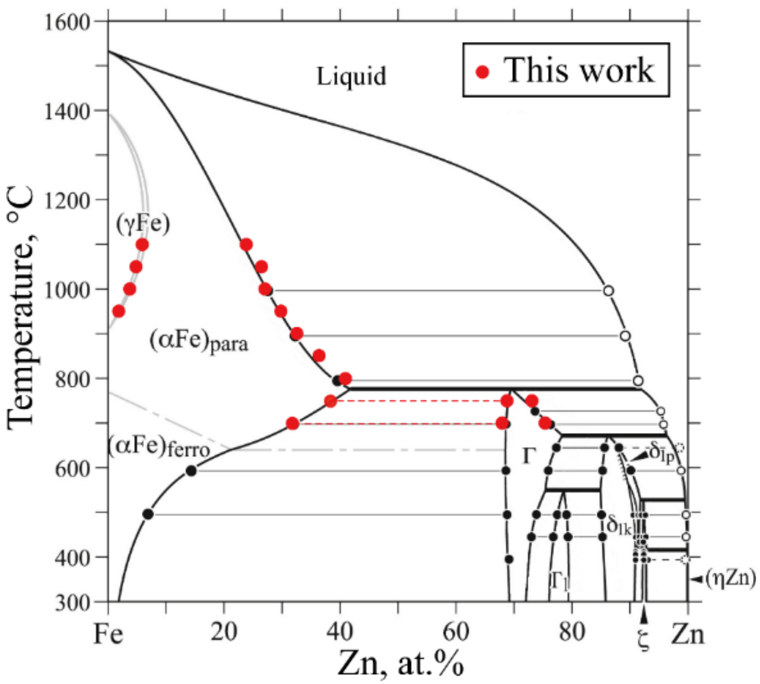


Task 6.0 – Thermodynamic Modeling

Collect more experimental data for future thermodynamic assessments to improve the current Al thermodynamic databases for designing AlFeSi-based alloys

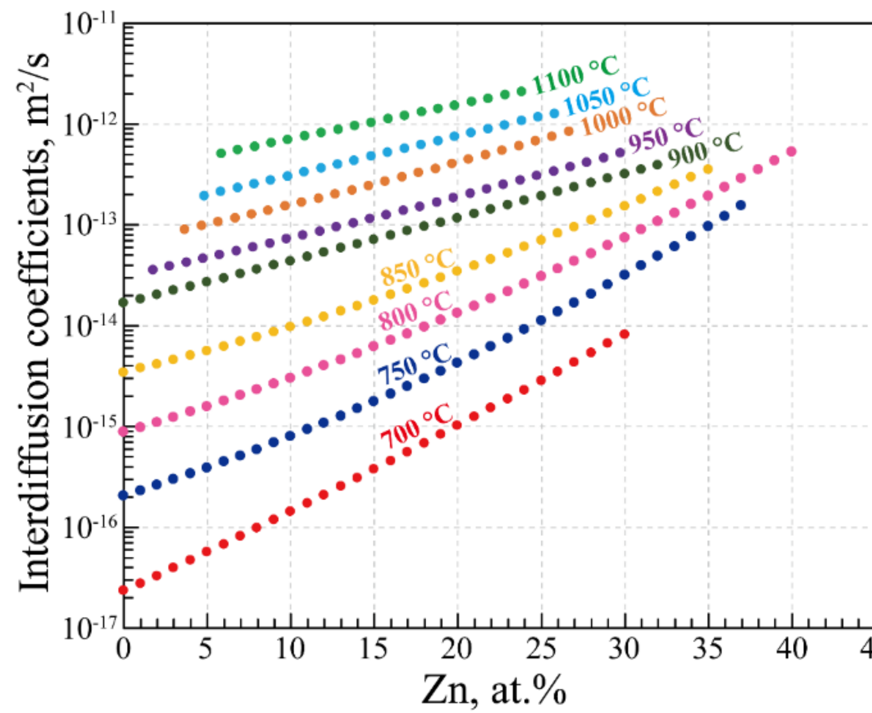
Phase equilibria and diffusion in the Fe-Zn system

Phase equilibrium data



Han, et al., J Alloys Compd. 737 (2018) 490-504.

Interdiffusion coefficients in αFe



Reviewer-Only Slides

Publications and Presentations

❑ Publications

- L.L. Zhu, S. Soto, R. Hennig, M. Manuel. Experimental investigation of the Al-Co-Fe phase diagram over the whole composition range, to be submitted.
- L.L. Zhu, S. Soto, R. Hennig, M. Manuel. Phase equilibria and diffusion coefficients in the Fe-Zn binary system, to be submitted.
- S. Soto, L.L. Zhu, B. Rijal, R. Hennig , M. Manuel. Experimental measurement of the phase boundary of the τ_{10}-$Al_4Fe_{1.7}Si$ intermetallic phase, to be submitted.
- L.L. Zhu, R. Hennig, M. Manuel. Experimental investigation of the Al-Cu-Fe phase diagram over the whole composition range, under preparation.

Publications and Presentations

❑ Presentations

- S. Soto, B. Rijal, L.L. Zhu, R. Hennig, M. Manuel. Microstructure Evolution of the High Temperature Intermetallic Phase τ_{10}-$Al_4Fe_{1.7}Si$. TMS 2018 Annual Meeting & Exhibition, March 13, 2018
- B. Rijal, R. Hennig, M. Manuel, S. Soto. Extension of the Stability Range of τ_{10} Phase in Al-Fe-Si Alloy: Cluster Expansion Approach. TMS 2018 Annual Meeting & Exhibition, March 13, 2018
- L.L. Zhu, S. Soto, R. Hennig, M. Manuel. Investigation of Al-Co-Fe and Al-Cu-Fe phase diagrams over the whole composition range. TMS 2019 Annual Meeting & Exhibition, March 13, 2019
- S. Soto, B. Rijal, L.L. Zhu, R. Hennig, M. Manuel. Solubility of Ni, Co and Mn in a lightweight Al-based high temperature intermetallic phase. TMS 2019 Annual Meeting & Exhibition, March 14, 2019

Critical Assumptions and Issues

- **The brittleness of the τ_{10}-$Al_4Fe_{1.7}Si$ intermetallic phase may present a limitation to its potential applications**
  - Grain boundary modification by minor alloying additions, such as B
- **The stability of the τ_{10}-$Al_4Fe_{1.7}Si$ intermetallic phase during printing heating cycles**
  - AlFeSiX alloys with a very high phase volume (up to 100%) had demonstrated to be stable at high temperatures (800 °C & 950 °C). Phase stability at a wider temperature range needs to be achieved.